



Carbon and oxygen isotopic survey of diagenetic carbonate deposits in the Agardhfjellet Formation (Upper Jurassic), Spitsbergen: preliminary results

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ABSTRACT: Diagenetic carbonate deposits (concretions, cementation bodies and cementstone bands) commonly occur in organic carbon-rich sequence of the Agardhfjellet Formation (Upper Jurassic) in Spitsbergen. They are dominated by dolomite/ankerite and siderite. These deposits originated as a result of displacive cementation of host sediment in a range of post-depositional environments, from shallow subsurface to deep-burial ones. Preliminary results of the carbon and oxygen isotopic survey of these deposits in southern Spitsbergen (Lågkollane, Ingebrigtsenbukta, Reinodden, and Lidfjellet sections) show the $\delta^{13}\text{C}$ values ranging between -13.0‰ and -1.8‰ VPDB, and the $\delta^{18}\text{O}$ values between -16.0‰ and -7.7‰ VPDB. These results suggest that the major stage of formation of the carbonate deposits occurred during burial diagenesis under increased temperature, most probably in late diagenetic to early catagenic environments. Carbonate carbon for mineral precipitation was derived from dissolution of skeletal carbonate and from thermal decomposition of organic matter.

Key words: Arctic, Spitsbergen, Agardhfjellet Formation (Jurassic), diagenetic carbonate deposits, carbon and oxygen isotopes.

Introduction

Organic carbon (OC)-rich, fine-grained facies of the Mesozoic sedimentary sequence in Spitsbergen (Harland 1977, Mørk *et al.* 1999) contain carbonate concretions, bands, and cementstone horizons originated in subsurface diagenetic environments. These carbonates record evolution of pore fluids during and after depositional history of the sequence, starting from shallow syngenetic environments (Krajewski and Luks 2003) down to deep-burial ones associated with Palaeogene subsidence and tectonic and thermal events in the region (Krajewski *et al.* 2001, Krajewski 2002). Therefore, they may potentially provide important in-



Fig. 1. Sketch map of Svalbard Archipelago showing location of studied sections of the Agardhfjellet Formation in Spitsbergen.

formation on stages of transformation and mineralization of organic carbon related to kerogen preservation and hydrocarbon generation in the sequence.

This paper presents preliminary results of a carbon and oxygen isotopic survey of diagenetic carbonate deposits in the Agardhfjellet Formation (Upper Jurassic),

which aid to understand processes that operated in burial environment of the OC-rich facies in Spitsbergen.

Geological setting

The Agardhfjellet Formation (Upper Jurassic) in Spitsbergen (Fig. 1) embraces a sequence of grey to black shales, mudstones and siltstones with subordinate sandstone intercalations that is very rich in organic carbon (Parker 1967, Birkenmajer 1975, 1980; Mørk and Bjorøy 1984, Krajewski 1989, Dypvik *et al.* 1991a). It was deposited in open marine shelf environment with periodic restricted water circulation (Dypvik 1980, 1985; Dypvik *et al.* 1991b). A prominent feature of the Agardhfjellet Formation is the presence of recurrent horizons containing diagenetic carbonate deposits dominated by dolomite/ankerite and siderite (Dypvik 1978, Krajewski 1992a, Bausch *et al.* 1998). They occur in a variety of forms, from concretions to sheet-like bodies, some of which attain several metres in size.

Most of the Agardhfjellet sequence in southern Spitsbergen consists of black muddy to silty shale up to approximately 180 m thick, which is referred to as the Ingebrigtsenbukta member (informal unit) in local lithostratigraphic scheme (Różycki 1959, Mørk *et al.* 1999). Four sections of the member were investigated for the purpose of this study (Fig. 2): (i) coastal cliffs at Lågkollane in Van Mijenfjorden (W Nathorst Land); (ii) coastal cliffs at Ingebrigtsenbukta in Van Keulenfjorden (NW Torell Land), (iii) coastal cliffs at Reinodden in Bellsund (NW Torell Land), and (iv) NW slope of Lidfjellet (NW Sørkapp Land). Detailed location of the sections as well as geological description of the sequence can be found in the following publications: Różycki (1959), Hjelle *et al.* (1986), Dallmann *et al.* (1990, 1993), Krajewski (1992a, b), Winsnes *et al.* (1993), Bausch *et al.* (1998), Dallmann (ed.) 1999, and Krajewski and Stempień-Sałek (2003).

Four distinct morphologic types of diagenetic carbonate deposits can be discerned in the Ingebrigtsenbukta member: (i) concretions; (ii) lensoidal cementation bodies; (iii) hummocky cementstone bands; (iv) compact cementstone bands and elongated bodies (Figs 3–5).

(I) The concretions usually show elliptical shapes and sharp boundaries towards the host sediment (Fig. 3). They vary in size from a few tens of centimetres up to several metres. The concretion bodies are composed of compact matrix dominated by fine carbonate cement. They are either massive or display variably cracked and/or brecciated central parts. The cracks are filled by coarse carbonate cement. Some concretions contain petrified wood fragments and/or fossils in their centres.

(II) The lensoidal cementation bodies embrace carbonate deposits that are controlled by the structure of the host sediment (Fig. 4A). They represent carbonate cementations of lenses of coarser-grained sediment (usually sand-sized) that occur

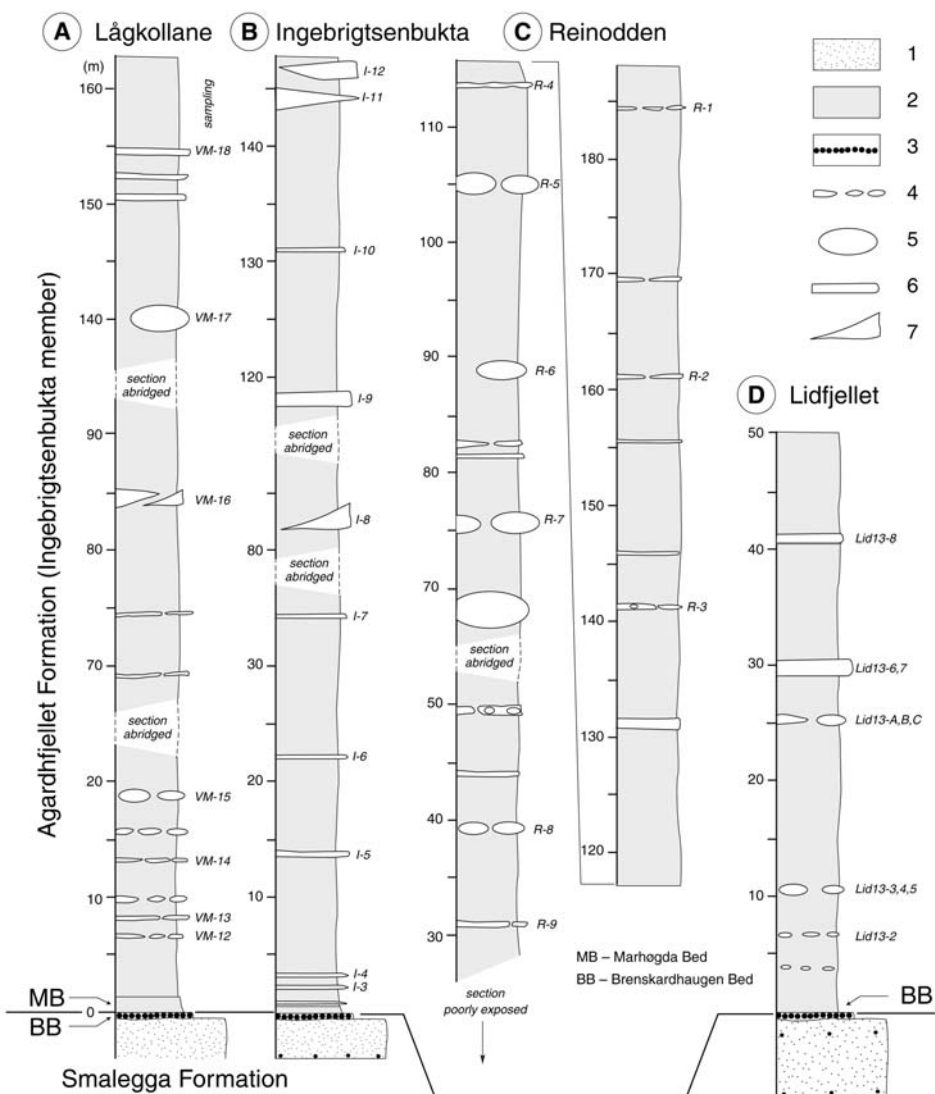


Fig. 2. Geological sections of the Agardhfjellet Formation (Ingebrigtsenbukta member) at Lågkollane (A), Ingebrigtsenbukta (B), Reinodden (C), and Lidfjellet (D), showing location of samples analyzed for the carbon and oxygen isotopic composition of diagenetic carbonate deposits. 1 – sandstone; 2 – OC-rich muddy to silty shale, siltstone, subordinate sandstone; 3 – phosphorite conglomerate; 4 – hummocky and concretionary cementstone bands; 5 – concretions; 6 – compact cementstone bands and elongated bodies; 7 – lensoidal cementation bodies.

in the black shale sequence. The bodies display relict lamination of the original sediment, and usually have diffuse boundaries towards the host. The matrix is dominated by fine-grained carbonate cement, similar to the one observed in concretions. Some of the bodies show more or less clearly defined zonation. Their

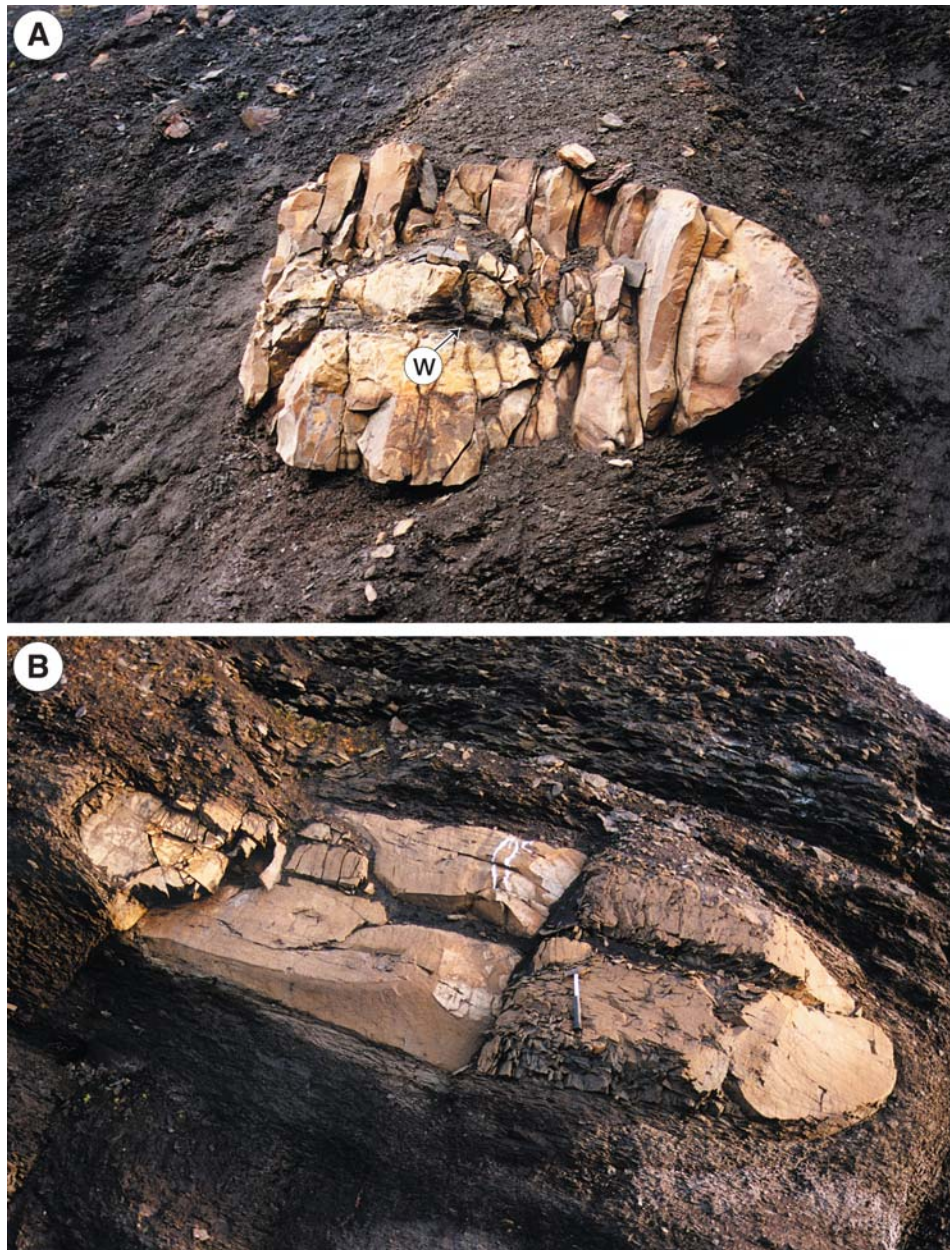


Fig. 3. **A.** Elliptical carbonate concretion dominated by dolomite/ankerite shows zoned internal structure. The concretion is developed around petrified wood fragment (w), and its central part is strongly brecciated. Horizontal axis of the concretion is 1.2 m long. Lidfjellet section. **B.** Massive carbonate concretion composed of a mixture of dolomite/ankerite and siderite. Hammer is 0.4 m long. Lågkollane section.

central parts are uncompacted and carbonate-rich, with the external parts being more clastic and mechanically oriented.

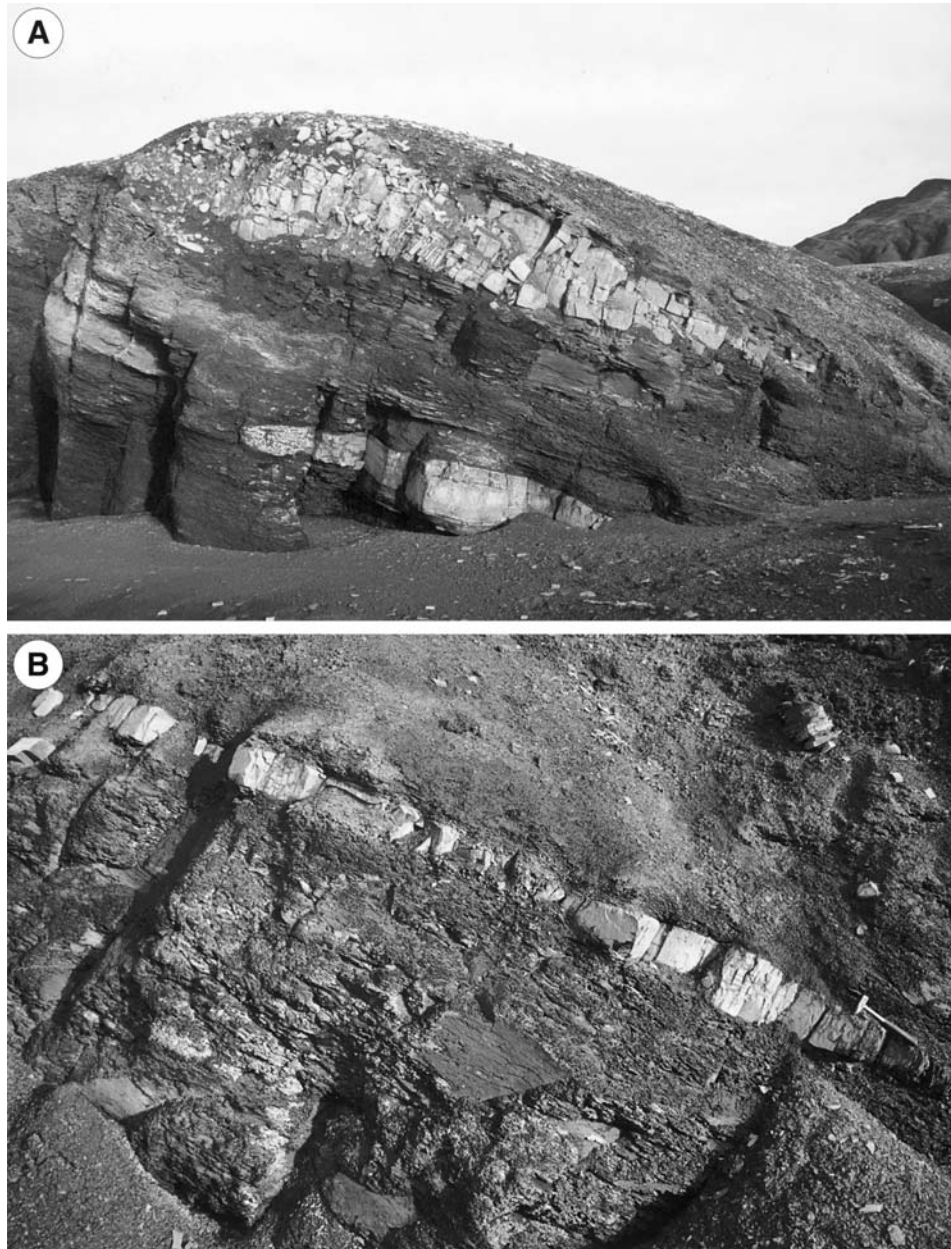


Fig. 4. **A.** Lensoidal carbonate cementation bodies occurring in black shale sequence show relicts of lamination of the original sediment. They are dominated by dolomite/ankerite cement. The lower body shows massive, carbonate-rich central part, and a marginal zone depleted in carbonate cement. The upper body is 8 m long. Lågkollane section. **B.** Hummocky cementstone band dominated by siderite in a sequence of black paper-shale. The band contains small carbonate concretions that are incorporated in laterally expanded cementstone. Hammer is 0.4 m long. Reinodden section.

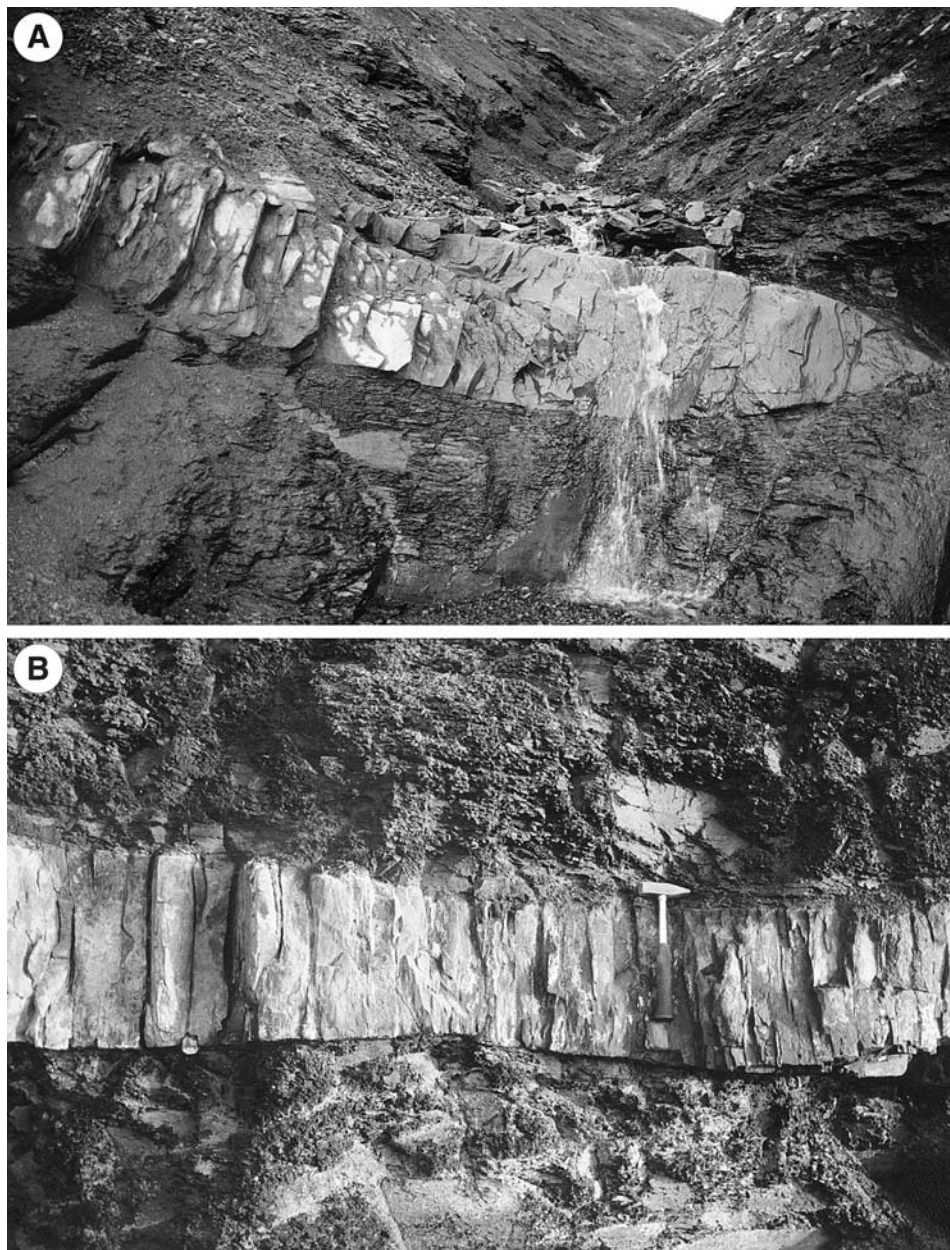


Fig. 5. **A.** Elongated cementstone body dominated by dolomite/ankerite in black shale sequence. The cementstone shows massive internal structure and tapered lateral termination. Maximum visible thickness of the body is 1.3 m. Lidfjellet section. **B.** Compact cementstone band dominated by dolomite/ankerite in a sequence of black paper-shale. Hammer is 0.4 m long. Lågkollane section.

(III) The hummocky cementstone bands embrace horizontally-oriented carbonate deposits that usually show multi-stage development (Fig. 4B). Incipient stages are represented by rows of small elliptical concretions (up to 0.5 m along horizontal axis) displaying massive or septarian internal structure. Advanced stages show carbonate cement overgrowths, preferentially along horizontal planes, that result in a characteristic, hummocky morphology of the mineral bodies. Composite horizons display changes in the content of carbonate cement from the concretions to their overgrowths, through the matrix is invariably fine-grained and structureless.

(IV) The compact cementstone bands embrace massive deposits that show tabular shapes and sharp boundaries towards the host sediment (Fig. 5). Tabular bodies are continuous laterally over a distance of several tens of metres. Their lateral terminations are either oblate or tapered, with or without noticeable warping of lamination of the host. The matrix is fine-grained and structureless throughout the analyzed sections.

The observed features of the carbonate deposits suggest their formation over a range of diagenetic environments as a result of displacive cementations of parts of the host sediment at various stages of its post-depositional compaction. Many carbonate bodies record long diagenetic history of formation, including stages of cement emplacement, induration, cracking, brecciation, and crack infilling.

Materials and methods

Thirty five horizons with diagenetic carbonate deposits in the Lågkollane, Ingebrigtsenbukta, Reinodden, and Lidfjellet sections of the Agardhfjellet Formation were sampled for the carbon and oxygen isotopic analysis (Fig. 2). One to three samples from each horizon were collected and analyzed separately (Table 1). All the analyzed samples represent compact matrix of carbonate bodies.

Mineral composition of the matrix was analyzed by means of X-ray diffraction (XRD). Samples were ground to $< 63 \mu\text{m}$ fraction using an agate mortar and pestle. Diffraction patterns were recorded on a SIGMA 2070 diffractometer using a curved position sensitive detector in the range $2\text{--}12^\circ 2\theta$ with $\text{CoK}\alpha$ radiation and 20 hour analysis time. DIFFRACTIONEL software v. 03/93 was used to process the obtained data. XRD data show that the carbonate bodies are composed of either dolomite/ankerite or siderite, or they contain various mixtures of the two (Fig. 4). Dolomite/ankerite volumetrically prevails over siderite in the sections studied. Pyrite and calcite are accessory diagenetic minerals revealed in some of these deposits.

The same samples and size fraction were analyzed for the carbon and oxygen isotopic composition of carbonate minerals. Carbonate CO_2 was produced from samples by reaction with anhydrous phosphoric acid ($d = 1.90 \text{ g cm}^{-3}$) under vacuum. In an attempt to discriminate between end members of carbonate mixtures, the samples were treated by a progressive acid extraction, with a time sequence of

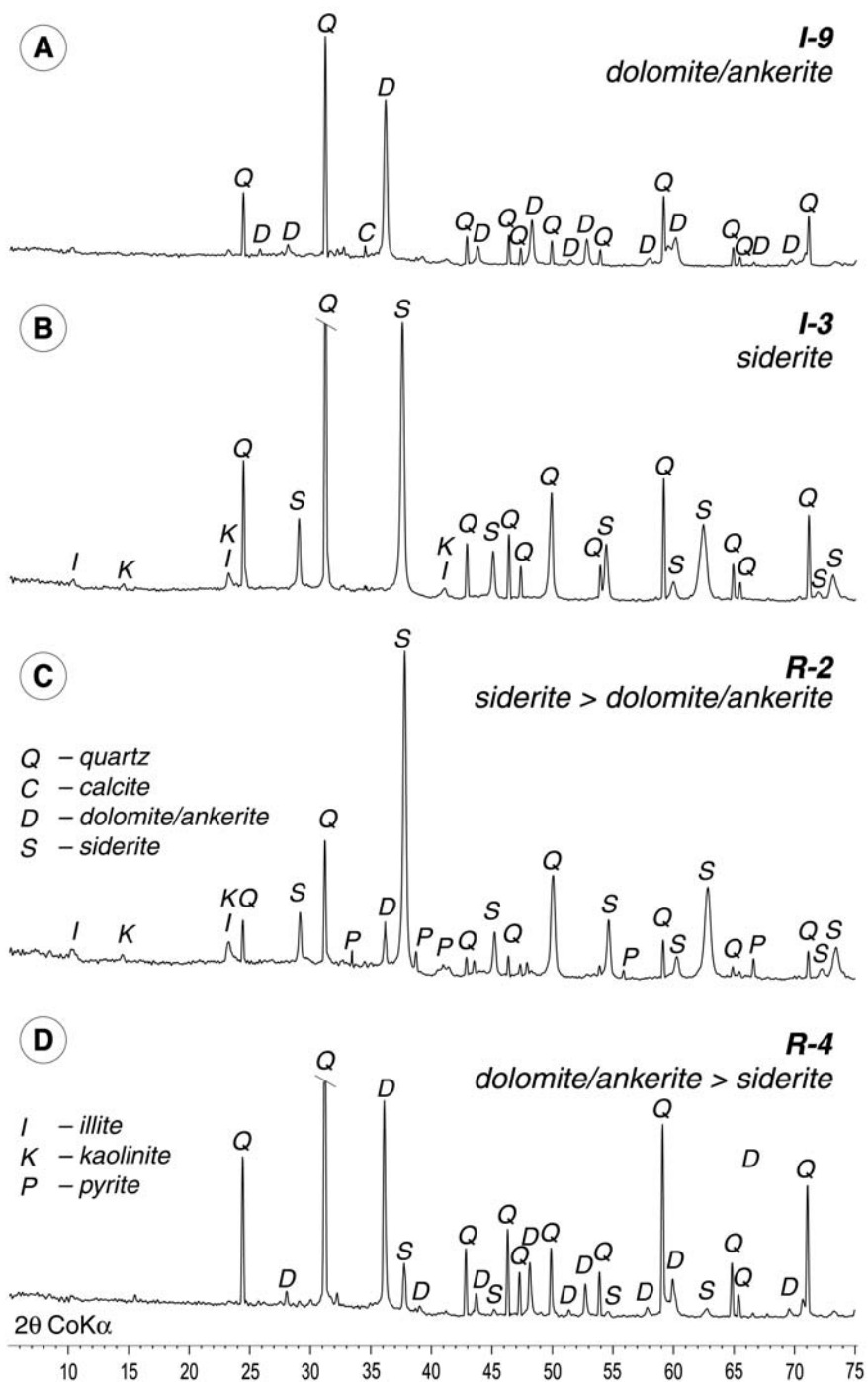


Fig. 6. X-ray diffractograms of selected samples of diagenetic carbonate deposits in the Agardhfjellet Formation showing their typical mineral composition. For location of samples see Fig. 2.

CO₂ collecting. CO₂ from samples containing calcite, dolomite/ankerite and siderite was collected after 24 hours of reaction at 25° C, and after 120 h at 50° C, with portions of gas discarded after 20 h at 25° C and after 48 h at 50° C. CO₂ from samples containing dolomite/ankerite and siderite was collected after 20 h at 25° C and after 120 h at 50° C, with a portion of gas discarded after 48 h at 50° C. CO₂ collected in these steps represents dolomite/ankerite and siderite, respectively. The procedure was verified by XRD (Figs 5, 6). Oxygen isotope fractionation factors $\alpha = 1.01098$ (25° C) and $\alpha = 1.01079$ (50° C) were used for dolomite/ankerite and siderite, respectively (Rosenbaum and Sheppard 1986, Carothers *et al.* 1988).

Isotopic ¹³C/¹²C and ¹⁸O/¹⁶O ratios were determined using a FINNIGAN MAT DELTA^{PLUS} spectrometer working in dual inlet mode with universal triple collector. The δ values were calculated relative to isotopic ratios of the international standard NBS 19. The results are expressed as $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ notations with respect to VPDB (Vienna Peedee Belemnite). Analytical reproducibility in laboratory was better than $\pm 0.05\text{‰}$ and $\pm 0.1\text{‰}$ for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$, respectively.

Results and discussion

The obtained $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values for dolomite/ankerite and siderite forming diagenetic carbonate deposits in the Agardhfjellet Formation are listed in Table 1. The $\delta^{13}\text{C}$ for dolomite/ankerite varies from -13.0‰ to -2.5‰ VPDB. The $\delta^{18}\text{O}$ varies from -13.8‰ to -7.8‰ VPDB. Mean values are -6‰ VPDB and -11‰ VPDB, respectively. The $\delta^{13}\text{C}$ for siderite varies from -11.3‰ to -1.8‰ VPDB. The $\delta^{18}\text{O}$ varies from -16.0‰ to -7.7‰ VPDB. Mean values are -6‰ VPDB and -10‰ VPDB, respectively.

These value ranges demonstrate that the two carbonate minerals have similar isotopic composition throughout the Agardhfjellet Formation, suggesting their origin under similar diagenetic conditions (Fig. 7). Mixed mineralogy of many analyzed carbonate bodies supports this interpretation (Fig. 4). However, the overlap of fields for dolomite/ankerite and siderite seen in Fig. 7 stands in contrast to a clear separation of these fields for the majority of diagenetic carbonate deposits in marine sequences (Mozley and Burns 1993). A likely explanation of this fact is that most of diagenetic deposits in the Agardhfjellet Formation originated in deep-burial environment, where the availability of iron, magnesium and calcium governed actual mineralogy of the precipitated carbonate, though its oxygen and carbon isotopic composition was controlled by a depth of formation in OC-rich sediment column.

Fig. 7 shows a rather good positive correlation between the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values for dolomite/ankerite and siderite. The calculated regression factor (r) is 0.67 and 0.70, respectively. The variation in $\delta^{13}\text{C}$ among the analyzed samples can be regarded as reflecting proportional variation in the supply of carbonate ion from

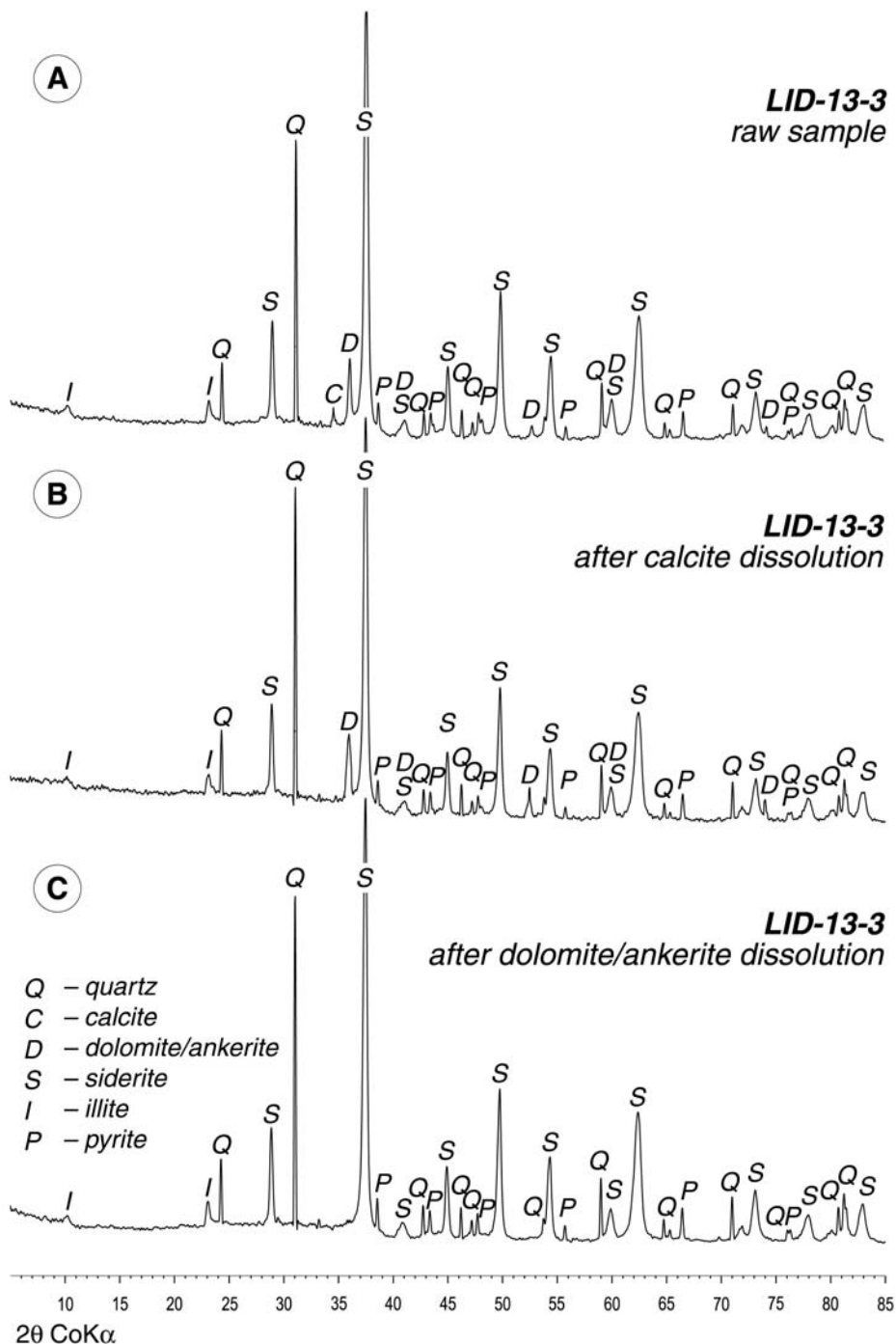


Fig. 7. X-ray diffractograms that verify a progressive acid extraction of CO₂ from sample LID-13-3 containing a mixture of calcite, dolomite/ankerite and siderite. For location of sample see Fig. 2.

Table 1
Isotopic composition of carbon and oxygen in carbonates forming diagenetic deposits in the Agardhfjellet Formation in Spitsbergen.

Sample		Dolomite/ankerite		Siderite	
		$\delta^{13}\text{C}$	$\delta^{18}\text{O}$	$\delta^{13}\text{C}$	$\delta^{18}\text{O}$
‰ (VPDB)					
Lågkollane	VM-12	-5.0	-10.0	-5.2	-9.9
	VM-13	-4.7	-10.1	-5.6	-10.5
	VM-14	-4.5	-10.2	-2.7	-9.8
	VM-15	-7.3	-10.9	-5.2	-10.6
	VM-16	-13.0	-12.7	-	-
	VM-17	-10.2	-12.9	-	-
	VM-18	-2.5	-8.4	-	-
Ingebrigtsenbukta	I-3	-	-	-9.8	-10.4
	I-4	-5.2	-9.4	-6.02	-9.5
	I-5	-3.0	-7.8	-1.8	-7.7
	I-6	-5.8	-10.9	-4.9	-9.4
	I-7	-5.8	-10.0	-6.0	-10.7
	I-9	-3.0	-10.0	-	-
	I-10	-6.7	-10.8	-	-
	I-11	-4.9	-10.0	-	-
	I-12	-9.1	-13.5	-	-
Reinodden	R-9	-10.0	-13.5	-	-
	haR-8	-8.3	-12.0	-	-
	R-7	-10.5	-12.7	-	-
	R-6	-7.4	-11.6	-	-
	R-5B	-9.7	-12.9	-	-
	R-5A	-7.7	-12.5	-	-
	R-4	-11.0	-12.9	-10.9	-12.8
	R-3	-11.6	-13.4	-9.3	-16.0
	R-2.1	-11.2	-11.2	-	-
	R-2.2	-11.2	-11.2	-11.3	-11.1
R-1	-5.8	-12.0	-	-	
Lidfjellet	LID-13-2	-3.5	-9.9	-	-
	LID-13-3.1	-3.7	-8.8	-	-
	LID-13-3.2	-3.7	-9.1	-3.9	-8.3
	LID-13-3.3	-6.2	-13.8	-	-
	LID-13-4	-4.1	-9.5	-4.1	-9.5
	LID-13-5	-4.2	-10.3	-4.0	-9.9
	LID-13-6	-4.4	-10.7	-	-
	LID-13-7.1	-3.3	-10.7	-	-
	LID-13-7.2	-3.3	-10.8	-	-
	LID-13-8.1	-7.3	-12.0	-6.5	-10.1
	LID-13-8.2	-7.4	-12.5	-	-
	LID-13-A.1	-3.4	-10.6	-	-
	LID-13-A.2	-5.1	-13.1	-	-
	LID-13-B.1	-5.4	-13.1	-	-
	LID-13-B.2	-4.8	-13.0	-	-
	LID-13-B.3	-3.4	-10.2	-	-
	LID-13-B.4	-5.2	-13.3	-	-
LID-13-C	-3.7	-9.4	-	-	

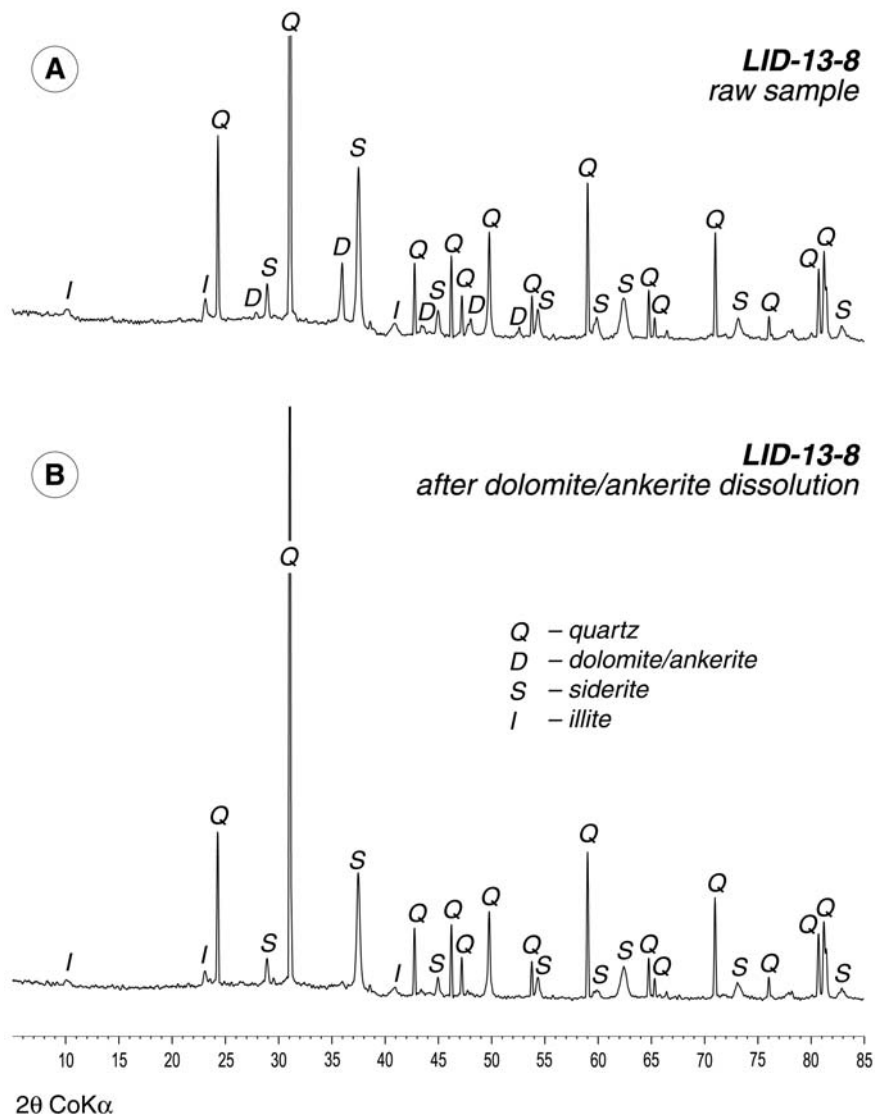


Fig. 8. X-ray diffractograms that verify a progressive acid extraction of CO₂ from sample LID-13-8 containing a mixture of dolomite/ankerite and siderite. For location of sample see Fig. 2.

isotopically different sources. The dominant sources in burial environment of the Agardhfjellet Formation were: (i) dissolution of mineral (skeletal) carbonate; and (ii) decomposition of organic matter. A palaeontologic studies show that vast majority of fossils in the formation occur in the form of moulds or imprints, without mineral skeletons preserved (Birkenmajer and Pugaczewska 1975, Kopik and Wierzbowski 1988). The exception is belemnite rostra (Ditchfield 1997, Ditch-

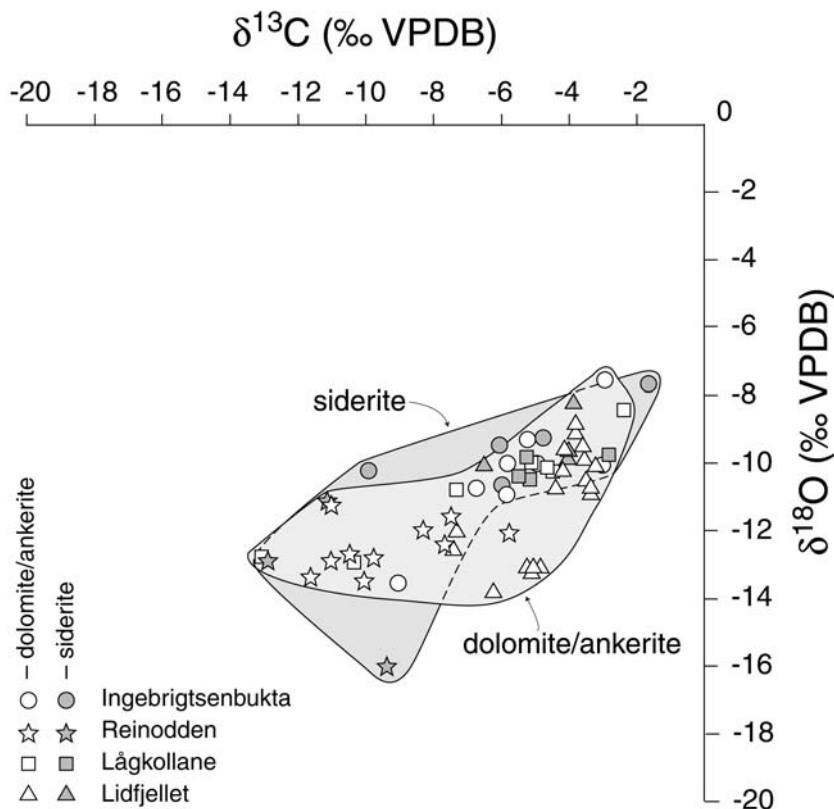


Fig. 9. Plot of $\delta^{13}\text{C}$ (‰ VPDB) versus $\delta^{18}\text{O}$ (‰ VPDB) of diagenetic carbonate deposits in the Agardhfjellet Formation in Spitsbergen.

field in Harland 1997, p. 381) that originally were composed of low-Mg calcite, though locally they also show traces of burial dissolution (Krajewski 2002). Organic geochemistry studies document a high content of organic carbon in the formation, reaching 12 wt. % (Mørk and Bjørø 1984). Kerogen of the Type II/III dominates the sequence, suggesting contribution of marine and land-derived organic matter (Bjørø and Vigran, 1980, Krajewski 1989). The recorded $\delta^{13}\text{C}$ values for the diagenetic deposits suggest that organic carbon provided less carbonate compared with the mineral one (Table 1). However, a clear decrease in $\delta^{13}\text{C}$, which reflects increasing contribution of the organic carbon source, is correlated with a consequent depletion in ^{18}O (Fig. 7). Similar isotopic trend has been revealed in burial ankerites forming replacement structures in the Marhøgda Bed at base of the Agardhfjellet Formation (Krajewski 2002).

The oxygen isotope composition can be used to calculate precipitation temperatures if the composition of precipitating fluids and equilibrium relationships controlling $\delta^{18}\text{O}$ are assumed (Krajewski *et al.* 2001, for discussion). Precipitation

temperatures calculated for dolomite/ankerite fall in a range between 80–140° C and 70–130° C (for assumed $\delta^{18}\text{O}_{\text{pore water}} = -1\text{‰ SMOW}$), depending on whether dolomite–water or ankerite–water isotopic fractionation equation is used (Land 1985, Fisher and Land 1986). Precipitation temperatures calculated for siderite fall in a range between 65–135° C, using the siderite–water isotopic fractionation equation of Carothers *et al.* (1988). These temperature ranges suggest formation of the carbonate deposits in a deep-burial environment. They also suggest that within this environment organic matter was preferentially decomposed due to thermal processes (decarboxylation of organic acids, kerogen breakdown). Increase in burial temperature was associated with accelerated production of biogenic carbonate to the pore fluids, which is consistent with the obtained $\delta^{13}\text{C}$ values. The apparent lack of methanogenic carbonate carbon within the diagenetic deposits seems to be a typical feature of the Mesozoic sequence in Spitsbergen, suggesting early kerogenization of organic matter (Krajewski *et al.* 2001, Krajewski 2002, Krajewski and Luks 2003).

Conclusions

Geological observations of diagenetic carbonate deposits in the Agardhfjellet Formation (Upper Jurassic) in Spitsbergen indicate that they originated over a range of post-depositional environments, from shallow subsurface to deep-burial ones. However, preliminary results of the carbon and oxygen isotopic survey of these deposits in the Ingebrigtsenbukta member in southern Spitsbergen suggest that the major stage of their formation occurred during burial diagenesis under increased temperature, most probably in late diagenetic to early catagenic environments. Carbonate carbon for mineral precipitation was derived from dissolution of skeletal carbonate and from thermal decomposition of organic matter.

Acknowledgements. — The field work in Spitsbergen (summer seasons 2001 and 2002) was supported by a research project No. 6PO4D04120 of the State Committee for Scientific Research. Logistic support of the Polish Polar Station in Hornsund and *M/S Horyzont II* is greatly appreciated. Tomek Nowicki and Bartek Luks helped the author during the field work. Bożena Łącka and Michał Kuźniarski performed the isotopic and X-ray analyses, respectively. The carbon and oxygen isotopic analysis was done at the Stable Isotope Laboratory of the Polish Academy of Sciences in Warszawa. The paper benefited from reviews by Krzysztof Birkenmajer, Michał Gruszczyński and Krzysztof Małkowski.

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Received 25 November 2003

Accepted 9 January 2004

